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ASSESSMENT OF AIR QUALITY IN BARCELONA BY PERSONAL MONITORING OF NONSMOKERS FOR RESPIRABLE SUSPENDED PARTICLES AND ENVIRONMENTAL TOBACCO SMOKE

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Personal monitoring over a 24-h period was performed using over 190 subjects divided into two distinct groups, one for housewives and one for office workers. Questionnaire surveys were conducted in addition to determinations for respirable suspended particles (RSP), environmental tobacco smoke (ETS) particles, nicotine, 3-ethenylpyridine, and saliva cotinine. The highest median levels of RSP, ETS particles, and nicotine were measured for subjects working in smoking workplaces. However, workers living in smoking households were more exposed outside working hours since more time was spent outside the workplace. These subjects, based upon median levels (90th percentile in parentheses), would be exposed each year to between 5.2 and 8.4 (26 and 40) cigarette equivalents (CE) outside the workplace compared with approximately 3.5 (13) CE at work. The lowest median levels were recorded for housewives living in nonsmoking households, equivalent to an exposure of approximately 1 CE per y. Subjective assessments of ETS exposure over the monitoring periods made by subjects on two separate occasions were considered to be consistent with measured concentrations. Subjects were not considered to have taken the length of time spent in any one environment into consideration, the ETS concentration rather than "overall exposure" having been assessed. Saliva cotinine measurements were used during the course of this investigation as a tool for determining misclassification of smoking status rather than a marker for ETS exposure. Using a cut-off level of 25 ng mL^{-1}, between 10.5% and 17.8% of the subjects were found to have misclassified themselves as nonsmokers, depending upon the criteria used.

INTRODUCTION

In a recent study performed by Sunyer et al. (1996), examining outdoor air pollution and mortality in Barcelona, the levels of black smoke, particles, and nitrogen dioxide were amongst the highest recorded in western Europe. This study was performed in accordance with the APHEA protocol (Katsouyanni et al. 1996) where fixed site monitoring was used for the measurement of particles, nitrogen dioxide, sulphur dioxide, and ozone with at least three sites per pollutant being used in most cases. Previous chemical analysis

Table 1. Cell categorization by home and workplace status – Barcelona.

Cell number	Study type	Smoking status	
		Home	Work
1	Single monitor	Smoking	-
2	Single monitor	Nonsmoking	-
3	Dual monitor	Smoking	Smoking
4	Dual monitor	Smoking	Nonsmoking
5	Dual monitor	Nonsmoking	Smoking
6	Dual monitor	Nonsmoking	Nonsmoking

of particles collected from outdoor air in Barcelona had shown approximately 50% of particulate matter was attributed to soil dust, a significant proportion to vehicle emissions (35%), and 1% contributed by industrial and heating emissions (Aceves and Grimalt 1993).

The present research in Barcelona was the second in a series of major European cities studied for air quality, again mainly focused on indoor air pollutants in homes and offices with specific reference to respirable suspended particles (RSP) and environmental tobacco smoke (ETS) particles. This study followed directly from the investigation of air quality in Stockholm using an identical protocol, with details of the methods, materials, and findings reported in a separate publication (Phillips et al. 1996).

The aim of the study was to determine personal exposures to RSP and ETS particles, the latter considered to be one of the major contributors to RSP in indoor air. Personal monitoring of nonsmokers was performed over a 24-h period combined with the self-reporting of activities using diaries and questionnaires. Saliva samples were taken for cotinine analysis to confirm nonsmoking status and estimate the rate at which smokers misclassify themselves as nonsmokers.

METHODS

Recruitment of subjects

The sample population for Barcelona was selected using the Mosaic classification system (Phillips et al. 1996), the 25 Mosaic types for Spain having been combined to be represented by 8 Mosaic groups. The sample selected was chosen in compliance with the following limitations:

1) All subjects to be living within 15 km from the city centre of Barcelona;

2) A third in each of the three age groups 20-34, 35-49, and 50-64; and,

3) Equal percentage distribution in Mosaic groups as for the population 15 km from the centre of the city.

The Mosaic classification enabled the comparison of the selected subjects with the Barcelona population using randomly selected telephone numbers from the files created using the above criteria. For Barcelona, the Mosaic group classification did not extend as far as the individual. Instead, a mixture of Mosaic groups was assigned to each individual in direct proportion with the distribution of Mosaic groups found within the specific area of residence. An aggregate of all the Mosaic group contributions from the sample population could then be compared with the actual distribution of Mosaic groups within the target area of Barcelona.

Initial telephone contact with subjects was carried out by Teleperformance, a large Opinion Research Bureau resident in Spain, although the operation was co-ordinated via their offices in Milan. Preliminary screening involved prospective volunteers being asked, "Are you 20 to 64 years of age and a nonsmoker?" If they responded affirmatively, they were asked if they would be prepared to participate in a general air quality survey. Suitable subjects were then further screened over the telephone to confirm their eligibility and to enable their assignment into one of six categories (Cells) for investigation (Table 1). Cells 1 and 2 were intended for housewives and therefore for subjects who did not work, and Cells 3 to 6 were for employed subjects, with office or non-industrial workers specifically targeted.

The monitoring session

Air samples were taken over a 24-h period either using a single personal monitor for the entire duration (single monitor study) or using two personal monitors sequentially over the same period (dual monitor study). The methodology employed for personal monitoring

has been described previously by these authors (Phillips et al. 1996) and consisted briefly of the following:

Initial visit to the study centre: Suitable volunteers were given an appointment to attend an information/training session organized at the centrally located Hotel Avenida Palace, in Barcelona. Here they were provided with full instructions regarding the use of monitoring equipment and the completion of study documentation. This included an instructional video in Spanish showing the correct method for operating the personal monitor. Subjects were required to complete a "first visit" questionnaire and provide a saliva sample prior to the monitoring period (pre-sample).

Single monitor study—housewife assessment: Non-working subjects recruited for participation in Cells 1 and 2 were provided with a single personal monitor and a diary/questionnaire for recording exposures and observations over the 24-h collection period.

Dual monitor study—home and work assessments: Working subjects recruited for participation in Cells 3 to 6 were provided with two personal monitors and diaries/questionnaires for recording exposures and observations whilst at work and for the remainder of the 24-h collection period. One monitor was worn at all times within the workplace, subjects having been encouraged to take lunch at their workstation wherever possible. A separate monitor was worn at all other times including travel to and from the workplace.

Final visit to the study centre: Both single monitor and dual monitor subjects were required to return their personal monitors and completed diaries and surveys to the study centre, where they answered a "last visit" questionnaire. They were also required to provide a second saliva sample (post-sample).

The personal monitor

Exposures were assessed using a personal monitor designed to collect RSP, ETS particles, and vapour phase components from the air close to the subject's breathing zone throughout a 24-h period (Phillips et al. 1996). RSP and ETS particles were collected using a Fluoropore membrane filter, and nicotine and 3-ethenylpyridine (3-EP) were collected by adsorption onto XAD-4 resin.

ANALYTICAL PROCEDURES

The analytical procedures performed during the course of this investigation have previously been reported and described in detail by these authors (Phillips et al. 1996) and comprised the following determinations:

1) RSP—performed using a gravimetric procedure.
2) ETS particles—estimated in three ways using high-performance liquid chromatography (HPLC) techniques and the application of predetermined factors. Estimates based upon solanesol content (SolPM), UV absorbance (UVPM), and fluorescence (FPM) of filter extracts were made. The factors for SolPM (44), UVPM (8.1), and FPM (44) were determined from controlled measurements using the six leading Spanish cigarette brand-types smoked in an environmental test chamber (Nelson et al. 1997).
3) Nicotine and 3-EP—performed using a capillary gas chromatographic procedure.
4) Saliva cotinine—performed using a radioimmunoassay procedure.

Limits of quantification (LOQ) for all analytes

Validated methods for the determination of UVPM, FPM, SolPM, nicotine, 3-EP, and cotinine were used. The limit of quantification (LOQ) for each analyte was determined as the lowest concentration for which precision and accuracy, both within each batch and between batches, did not exceed ± 20%. The LOQ for the measurement of RSP was determined from the weight change of filter blanks prepared throughout the study. A value of the mean plus two standard deviations for these measurements was used.

With the exception of cotinine, Table 2 presents analytical LOQs in terms of air concentrations for each method based upon sample collection periods, assuming filter and sorbent tube flow rates of exactly 1.72 and 0.8 L min^{-1}, respectively. Where levels were found below the LOQ, a value of one-half of the LOQ was used for the data analysis, the same procedure having been used previously by these authors (Phillips et al. 1996).

RECRUITMENT OF SUBJECTS

The recruitment of subjects for study participation in Barcelona proved difficult for a number of reasons. Recruitment of subjects into Cells 4 and 6 of the dual monitor study, requiring the subject to be working in a nonsmoking workplace, was extremely difficult since

Table 2. Limits of quantification for the analytical methods according to collection period – Barcelona.

Analyte	Collection period		
	24 h	15.6 h *	7.8 h **
Respirable suspenced particles (RSP)	9.90 μg m^{-3}	15.2 μg m^{-3}	30.5 μg m^{-3}
ETS particles measured by UV (UVPM)	0.49 μg m^{-3}	0.75 μg m^{-3}	1.51 μg m^{-3}
ETS particles measured by fluorescence (FPM)	0.11 μg m^{-3}	0.17 μg m^{-3}	0.34 μg m^{-3}
ETS particles measured by solanesol (SolPM)	0.27 μg m^{-3}	0.41 μg m^{-3}	0.82 μg m^{-3}
Nicotine	0.09 μg m^{-3}	0.13 μg m^{-3}	0.27 μg m^{-3}
3-Ethenylpyridine	0.09 μg m^{-3}	0.13 μg m^{-3}	0.27 μg m^{-3}
Saliva cotinine	1.0 ng mL^{-1}	-	-

* Mean time spent outside the workplace for working subjects in Barcelona.
** Mean time spent in the workplace for working subjects in Barcelona.

Table 3. Age and sex distribution for study subjects – Barcelona.

Cell	Sex		Age range (y)					Overall total
	Male	Female	< 20	20 - 34	35 - 49	50 - 64	> 64	
1		43		7	15	21		43
2	2	40		4	18	19	1	42
3	10	15	1	8	8	8		25
4		3			2	1		3
5	22	14		12	19	5		36
6	4	1		1	4			5
Single monitor total	2	83		11	33	40	1	85
Dual monitor total	36	33	1	21	33	14		69
Overall total	38	116	1	32	66	54	1	154

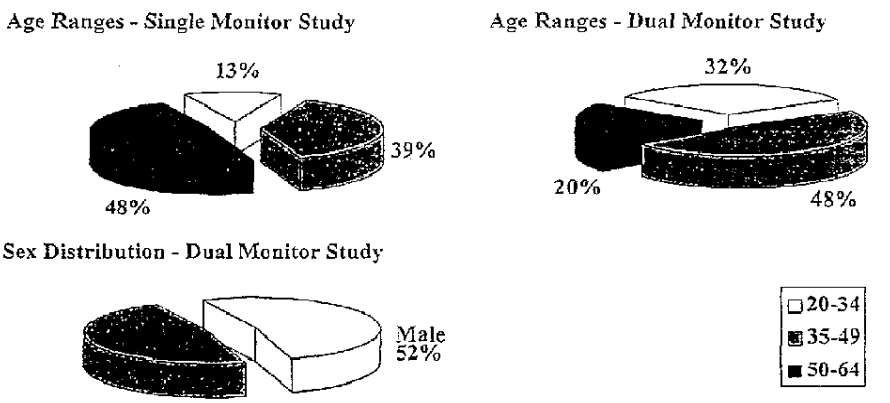

Fig. 1. Age range and sex distributions for recruited subjects in Barcelona.

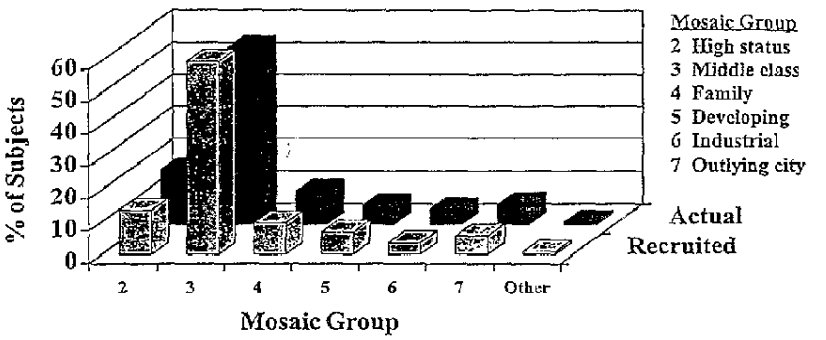

Fig. 2. Distribution of subject lifestyles – Barcelona.

smoking was prevalent in most office environments. In addition, recruitment sessions performed at the study centre suffered from mass walkouts. On a number of occasions, as many as 12 volunteers left mid-session, mainly due to a reluctance to complete the comprehensive array of study questionnaires. Volunteer losses were also encountered after the 24-h monitoring period when it was discovered that some subjects had failed to switch on their monitors. In order to counter these unforeseen losses, the telephone recruitment rate was increased and the study period extended.

Of the 197 volunteers claiming to be nonsmokers that were recruited for this study, 16 were excluded because they admitted to being smokers on the "first visit" questionnaire, although they had claimed nonsmoking status in their initial telephone contact. This change in response may be due to subjects being asked at two different points in time or be attributable to additional information being provided during the video presentation, thus making the subjects reconsider their position.

A further 27 subjects were excluded from the study, 19 of which were not admissible due to their saliva cotinine levels being above the selected threshold (25 ng mL^{-1}) for nonsmokers and a further 8 subjects failed to collect their samples. Of the remaining 154 subjects who successfully completed the study, age and sex distributions are presented in Table 3 and Fig. 1.

Two subjects falling outside the specified age ranges, one younger than 20 and one older than 64, were not excluded from the study. In addition, two male subjects recruited into Cell 2 were included in the study in order to obtain much needed data on ETS exposure outside the workplace.

Age and sex distribution

Figure 1 shows the distribution of males and females recruited for the dual monitor study to be close to that planned (50% per sex). The distribution of ages within the single monitor study was biased towards the older population with 48% of subjects aged over 50 y and only 13% of subjects being younger than 35 y of age. For the dual monitor study, the largest group comprised subjects within the 35 to 49 y age range with a corresponding shortfall in the oldest age group. The aim was to recruit equal numbers of volunteers into each age range.

Geodemographic distribution

The study was designed to have participating subjects representative of the population of Barcelona. Figure 2 shows that the participants on the study closely resemble the population as expressed in Mosaic lifestyles. Therefore, there is no reason to believe the study has attracted a distribution of people significantly different from that representing the total population of Barcelona. The population sectors contained within each Mosaic group are presented in Table 4.

Occupations

The subjects were restricted to a choice of 12 occupations from which to select and provide their answers

Table 4. Population sectors contained within Mosaic groups – Barcelona.

Mosaic group	Population sectors
1	Areas of tourism
2	High status areas
3	Middle class areas
4	Family areas
5	Developing areas
6	Industrial areas
7	Outlying city areas
8	Depressed areas

Table 5. Occupations of recruited subjects – Barcelona.

Occupation	Number of responses
Administration	28
Building trade	1
Education	6
Engineering	5
Government	1
Legal	3
Medical	4
Other	16
Retail	1
Science	2
Supply industry	1
Transport	1
Total	69

Table 6. Maximum measured concentrations (½ hourly) of airborne pollutants for Barcelona throughout the study period (all stations) January 1995.

Analyte	Concentration
Particles	168 $\mu g\ m^{-3}$
NO_2	1266 $\mu g\ m^{-3}$
SO_2	162 $\mu g\ m^{-3}$
O_3	102 $\mu g\ m^{-3}$
CO	16.9 $mg\ m^{-3}$

on the last visit questionnaire. Table 5 lists these occupations and the answers that were provided by the 69 subjects wearing the workplace monitor on this study.

MISCLASSIFICATION OR MISREPORTING OF SMOKING STATUS

This study did not set out to investigate misreporting of smoking status in any detail. Another study, run concurrently with this one, focused on misclassification and smoking history and will be the subject of a detailed publication in the future.

To determine whether subjects had misreported their smoking status, a saliva cotinine threshold limit of 25 ng mL^{-1} was used, as described previously by these authors (Phillips et al. 1996). Of the 16 subjects admitting to being smokers by means of questionnaire responses, 2 were not identified as smokers by their saliva cotinine measurements. This may demonstrate that saliva cotinine measurements can fail to distinguish between nonsmokers and occasional smokers who may not have smoked for several days.

Depending upon the criteria used, which included responses to questionnaires, the rate at which recruited subjects misreported their smoking status ranged from 10.5% (19 of 181) to 17.8% (35 of 197).

WEATHER CONDITIONS DURING THE STUDY

Weather and other pollutants information

Detailed information about the weather conditions and the levels of certain airborne pollutants during the course of the study were obtained from the local environmental offices in Barcelona. The study was carried out during January 1995 with daily mean temperatures over this period varying from a minimum of 7.4°C to a maximum of 15.7°C. A maximum daily rainfall of 50 mm was recorded, with rain falling on three days of the study period. Mean daily wind speeds varied between 1.6 and 6.2 m s^{-1} and maximum and minimum relative humidities of 90 and 54%, respectively, were recorded.

Concentrations of airborne pollutants, namely particles, NO_2, SO_2, O_3, and CO were measured during the study period at a number of monitoring stations situated around the Barcelona area. The maximum concentrations measured for each pollutant, based on mean half-hourly measurements at any monitoring station, are presented in Table 6. Variations in daily minimum, maximum, and mean concentrations of NO_2 over the study period are presented in Fig. 3. The air quality bandings used in the UK to describe air quality are also depicted in this figure. On at least 4 d of the study period, mean nitrogen dioxide levels were in excess of 100 $\mu g\ m^{-3}$, when the air quality could be described as being poor.

At this stage, no attempt has been made to correlate personal monitoring measurements with weather conditions by, for example, modeling temperature and relative humidity fluctuations with relative analyte concentrations.

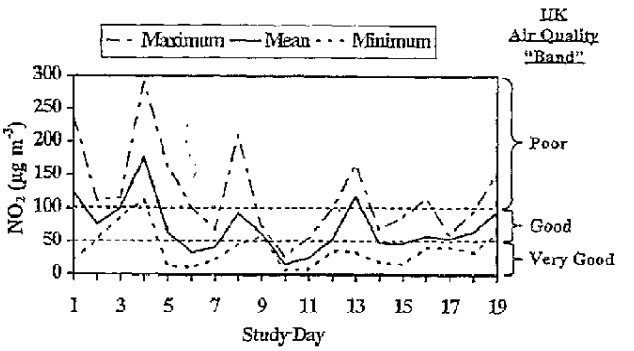

Fig. 3. Concentrations of atmospheric nitrogen dioxide – Barcelona.

RESULTS AND DISCUSSION

Comparison of overall exposures between housewives and working subjects

Because the results from this study are not normally distributed, median values for reporting RSP and ETS exposures were used. Additionally, in this publication, the arithmetic and geometric means for each concentration data set have been quoted together with the upper (90th percentile) and lower (10th percentile) decile values.

ETS particle concentrations measured using SolPM, a tobacco specific marker, have primarily been used throughout this study for the estimation of ETS exposure. However, on a number of occasions, subjects were found to be exposed to quantifiable concentrations of nicotine where measured SolPM concentrations were below the LOQ. This may be attributable to the presence of nicotine in areas where smoking had previously taken place, ETS particles being absent since smoking was not actively taking place at the time of measurement. Where high concentrations of ETS particles were quantified, levels determined using SolPM were higher than those determined using either UVPM or FPM. This is in contradiction to the expected trend UVPM > FPM > SolPM, the highest levels expected from the least specific method of determination (UVPM). This may be attributable to the use of predetermined factors in the calculation of ETS particle concentrations from ultraviolet, fluorescence, and solanesol measurements, which were determined using the top six selling cigarette brand-types in Spain. The presence of cigar or pipe tobacco smoke during sampling, for example, could significantly affect the solanesol content of the ETS particulate phase and, using cigarette based factors, may result in an overestimate of the levels present. On five occasions, estimated SolPM concentrations actually exceeded those for RSP, further indicating that ETS particle contributions may be overestimated especially at higher concentrations using SolPM measurements. Although ETS particle concentrations estimated using solanesol measurements are currently considered to give the closest representation of actual levels present, in light of these recent findings, it may be appropriate to consider FPM values alongside SolPM values. For ease of comparison with previous publications, nicotine results have been reported in this study.

Tables 7 and 8 show the summary analytical data for all subjects from the single monitor and dual monitor studies, respectively. Taking a combination of Cells 1 and 2, smoking and nonsmoking homes, the median RSP concentration for housewives was 57 $\mu g\ m^{-3}$ with an ETS particle contribution of 4.3 $\mu g\ m^{-3}$ based upon SolPM and 6.1 $\mu g\ m^{-3}$ based upon FPM measurements. Similar median levels were apparent outside the workplace in the dual monitor study, where RSP was 57 $\mu g\ m^{-3}$ with ETS particle contributions of 4.8 $\mu g\ m^{-3}$ and 7.4 $\mu g\ m^{-3}$ based upon SolPM and FPM measurements, respectively. Therefore, using SolPM measurements, ETS particles contribute between 7.5% and 8.4% (10.7% and 13.0% for FPM) of RSP outside of the workplace. In the workplace, the median exposure level for RSP was 89 $\mu g\ m^{-3}$ with an ETS particle contribution (SolPM) of 35 $\mu g\ m^{-3}$, equivalent to 39% of the particulate total. This may be an overestimate since the equivalent value based on FPM measurements was 27 $\mu g\ m^{-3}$, representing 30% of collected RSP.

Table 7. Summary analytical statistics for housewives from smoking and nonsmoking homes – Barcelona.

Analyte		Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
Pre-cotinine	(ng mL^{-1})	0.50	4.4	6.0	1.1	0.50	75
Post-cotinine	(ng mL^{-1})	0.50	3.4	4.3	0.94	0.50	73
RSP	(μg m^{-3})	32	139	73	61	57	83
UVPM	(μg m^{-3})	2.0	40	16	8.6	8.6	82
FPM	(μg m^{-3})	1.7	28	13	6.8	6.1	75
SolPM	(μg m^{-3})	0.29	54	17	3.6	4.3	82
Nicotine	(μg m^{-3})	0.05	2.4	0.86	0.33	0.22	83
3-Ethenylpyridine	(μg m^{-3})	0.04	1.8	0.54	0.24	0.21	83

Table 8. Summary analytical statistics for working subjects from all environments - Barcelona.

Analyte		Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
Pre-cotinine	(ng mL^{-1})	0.50	5.6	2.3	1.4	1.5	58
Post-cotinine	(ng mL^{-1})	0.50	3.9	2.1	1.4	1.5	55
			Home monitor				
RSP	(μg m^{-3})	29	144	74	59	57	68
UVPM	(μg m^{-3})	2.3	40	18	9.4	7.5	68
FPM	(μg m^{-3})	2.2	41	18	9.2	7.4	68
SolPM	(μg m^{-3})	0.69	59	21	5.5	4.8	68
Nicotine	(μg m^{-3})	0.07	2.9	1.0	0.36	0.29	69
3-Ethenylpyridine	(μg m^{-3})	0.06	1.7	0.55	0.25	0.21	69
			Work monitor				
RSP	(μg m^{-3})	42	182	106	88	89	68
UVPM	(μg m^{-3})	5.4	86	42	23	27	68
FPM	(μg m^{-3})	5.2	99	44	24	27	68
SolPM	(μg m^{-3})	1.1	128	58	18	35	68
Nicotine	(μg m^{-3})	0.35	8.5	3.7	2.0	2.2	69
3-Ethenylpyridine	(μg m^{-3})	0.14	2.8	1.4	0.86	1.0	69
			Total *				
RSP	(μg m^{-3})	42	135	84	74	71	68
UVPM	(μg m^{-3})	5.0	53	26	18	21	68
FPM	(μg m^{-3})	5.5	55	27	19	21	68
SolPM	(μg m^{-3})	2.4	81	33	17	22	68
Nicotine	(μg m^{-3})	0.31	3.8	1.9	1.2	1.2	69
3-Ethenylpyridine	(μg m^{-3})	0.20	1.9	0.81	0.58	0.57	69

* Values calculated as a time weighted average of exposure concentrations determined both inside and outside the workplace for each subject.

Overall median concentrations of RSP and ETS particles in the dual monitor study, combining Cells 3, 4, 5, and 6 using a time weighted combination of exposures outside the workplace and at work for each participant over the 24-h collection period, were 71 μg m^{-3} and 22 μg m^{-3} (based on SolPM), respectively. Using UVPM and FPM measurements, a median value of 21 μg m^{-3} was obtained, which provided near identical exposure concentration information.

Figure 4 shows the actual distribution of SolPM concentrations for the single monitor and dual monitor studies. The spread of concentrations measured outside

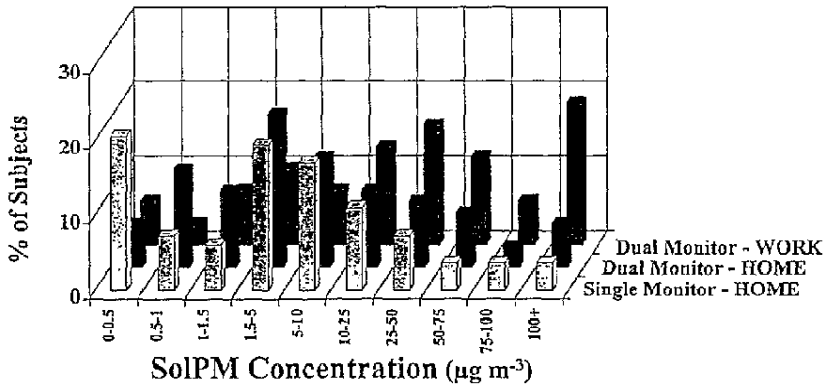

Fig. 4. Distribution of ETS particle concentrations. All subjects – Barcelona.

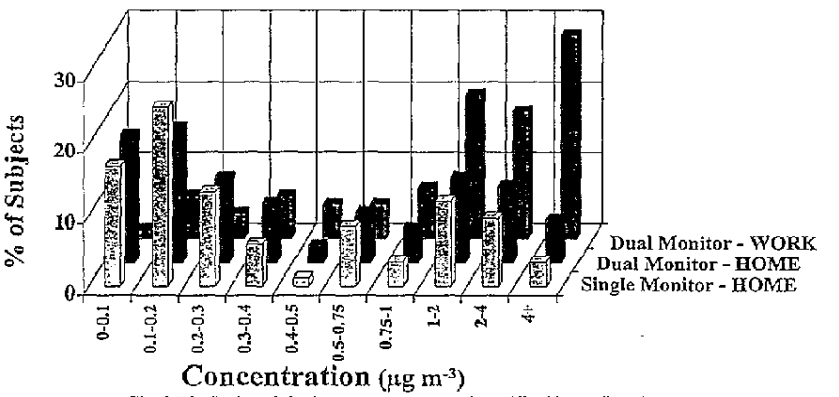

Fig. 5. Distribution of nicotine exposure concentrations. All subjects – Barcelona.

the workplace during the dual monitor study was comparable with those measured for subjects taking part in the single monitor study with 29.4 and 34.1% of concentrations, respectively, falling between 0 and 1.5 µg m^{-3} and the majority (54.9 and 54.4%, respectively) falling between 1.5 and 50 µg m^{-3}. In the workplace measured exposure concentrations were higher, with more than 73% of subjects having been exposed to levels exceeding 10 µg m^{-3} compared with around 30% of subjects for outside the workplace. More than 25% of these subjects at work were exposed to ETS particle concentrations in excess of 100 µg m^{-3}.

The distribution of nicotine exposure concentrations (Fig. 5) followed the same pattern as ETS particle measurements, with a close agreement between single and dual monitor home assessments. A larger proportion of higher levels was determined for the workplace portion of the dual monitor study.

The median exposure values for the single monitor study reported in Table 7 are significantly higher than the LOQs of the methods used, with RSP, SolPM, and nicotine levels equivalent to approximately 6, 16, and 2 times their respective LOQs. Subjective assessments made immediately after this sampling period, documented as part of the home pump survey, revealed that

Subjective ETS assessment

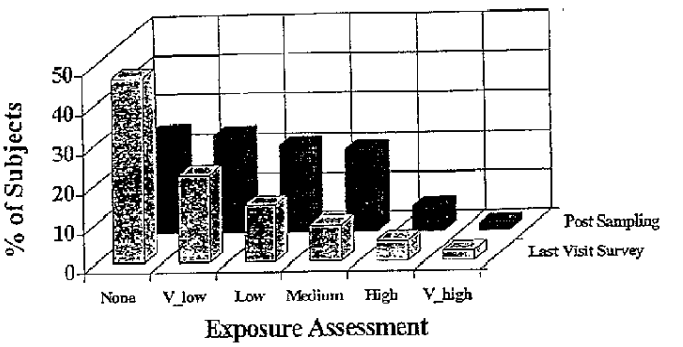

Fig. 6. Subjective assessment of ETS exposure. Single monitor study – Barcelona.

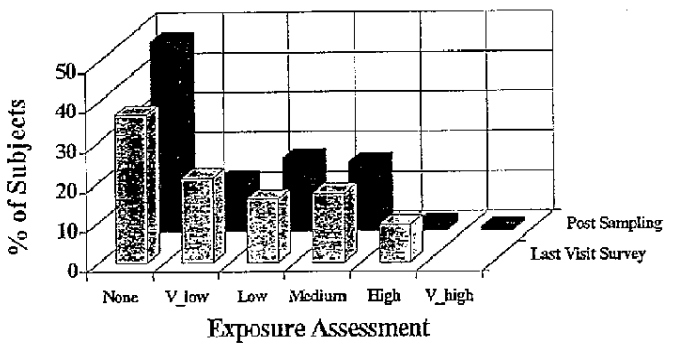

Fig. 7. Subjective assessment of ETS exposure. Dual monitor study (home) – Barcelona.

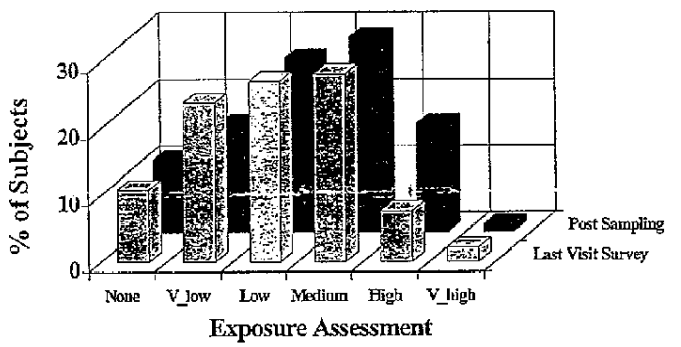

Fig. 8. Subjective assessment of ETS exposure. Dual monitor study (work) – Barcelona.

Table 9. Summary analytical statistics for all housewives by smoking environment – Barcelona.

Analyte		Cell *	Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
Pre-cotinine	(ng mL^{-1})	1	0.50	6.1	11	1.7	1.4	39
		2	0.50	1.7	1.0	0.68	0.50	36
Post-cotinine	(ng mL^{-1})	1	0.50	5.6	8.0	1.4	1.1	35
		2	0.50	1.8	0.95	0.65	0.50	38
RSP	(μg m^{-3})	1	35	155	82	70	63	43
		2	30	90	63	53	51	40
UVPM	(μg m^{-3})	1	4.5	60	23	15	15	42
		2	1.8	12	7.7	4.8	4.7	40
FPM	(μg m^{-3})	1	2.3	46	19	11	13	35
		2	1.7	11	7.1	4.4	4.4	40
SolPM	(μg m^{-3})	1	1.4	88	28	11	11	42
		2	0.13	8.1	5.6	1.2	1.0	40
Nicotine	(μg m^{-3})	1	0.13	2.8	1.4	0.72	0.74	43
		2	0.04	0.46	0.33	0.14	0.11	40
3-Ethenylpyridine	(μg m^{-3})	1	0.11	2.0	0.84	0.48	0.60	43
		2	0.04	0.33	0.23	0.12	0.12	40

* Cell 1 - smoking household; Cell 2 - nonsmoking household.

about 50% of subjects considered their ETS exposure to be "none" or "very low", approximately 40% considered their exposure to be "low" or "medium" and the remainder (about 7%) documented their exposure as "high" or "very high". These subjective assessments were performed again on returning the monitors to the study centre (last visit survey) and the spread of responses to the above criteria was approximately 69%, 23%, and 8%, respectively. These subjective assessments are depicted in Fig. 6.

For the dual monitor study, similar assessments were made by questionnaire for both the home and work environments (Figs. 7 and 8). In the "home pump survey" (last visit survey in parentheses) for the home portion, approximately 58% (61%) claimed that their exposure was "none" or "low", approximately 33% (36%) considered their exposure to be "low" or "medium", and the remaining 9% (3%) recorded their exposure as "high" or "very high". These subjective assessments made by working subjects outside the workplace were comparable with those made by housewives participating in the single monitor study. The distribution of analyte concentrations for nonworking subjects and working subjects outside the workplace were also comparable, reflecting their subjective assessments of ETS exposure. For the work portion, subjective assessment of ETS exposure was much higher than for the home portion. Post sampling responses, entered as part of the "work pump survey", indicated that approximately 27% of subjects considered their exposure to be "none" or "very low", around 55% considered their exposure to be "low" or "medium", and the remainder (approximately 17%) considered their exposure to be "high" or "very high". Corresponding subjective assessments made on the last visit survey were approximately 35%, 55%, and 10%, respectively, for the aforementioned categories. Actual measured exposure concentrations for the work portion were consistent with this subjective assessment, median levels being at least 7 times greater, based upon SolPM and nicotine, than for the home portion. This data indicates that subjects were capable of determining their degree of exposure in a particular environment, in proportion with the prevailing concentration of ETS present.

Housewife study

Differences between smoking and nonsmoking households: Summary analytical results by cell are presented in Table 9. Subjects living in smoking households were found to have higher median exposures to RSP (63 μg m^{-3}), ETS particles (11 μg m^{-3}), and nicotine (0.7 μg m^{-3}) than those living in nonsmoking households (51 μg m^{-3}, 1.0 μg m^{-3}, and 0.11 μg m^{-3}, respectively). Median pre- and post-cotinine saliva levels were also higher for the subjects living in smoking than in nonsmoking homes (pre- 1.4 ng mL^{-1}

Table 10. Calculated annual exposures to RSP, ETS particles, and nicotine for housewives – Barcelona.

Environment	Annual exposure (mg) RSP	ETS particles*	Nicotine	Cigarette equivalents†
	Median levels			
Smoking home	359	63	4.2	4.2
Nonsmoking home	290	5.7	0.63	0.4 - 0.6
	Upper decile levels			
Smoking home	883	501	16	16 - 33
Nonsmoking home	512	46	2.6	2.6 - 3.1

* Estimated using solanesol measurements (SolPM).
† Calculated using both ETS particle and nicotine measurements.

vs 0.5 ng mL^{-1}; post- 1.1 ng mL^{-1} vs 0.5 ng mL^{-1}). These levels found for subjects living in smoking homes are similar to those found by Phillips et al. (1994) for British subjects living with smoking partners and are approximately twice the levels found for subjects in Stockholm living in smoking homes (Phillips et al. 1996).

The median values found in this study of below 2 ng mL^{-1} are also comparable with serum cotinine levels determined as part of the Third National Health and Nutrition Examination Survey (NHANES III) reported by Pirkle et al. (1996). That survey reported a median serum cotinine level of 0.526 ng mL^{-1} for subjects reporting any ETS exposure either at home or at work. For ETS exposure in the home only, a geometric mean of 0.7 ng mL^{-1} was reported.

In Barcelona, geometric means of 1.4 ng mL^{-1} and 0.65 ng mL^{-1} for post-monitoring saliva cotinine measurements were apparent for housewives living in smoking and nonsmoking households, respectively. Serum and saliva measurements are considered to provide equivalent information regarding cotinine disposition in the body (Jarvis et al. 1987; Curvall et al. 1990).

The median exposure concentrations of ETS particles and nicotine for housewives living in smoking households were between 6 and 10 times higher than for those living in nonsmoking households. Corresponding median RSP levels were only 1.2 times higher in smoking homes than in nonsmoking homes. This is comparable with Guerin et al.'s (1992) summary of field studies where they indicate that RSP in smoking locations are typically a factor of 1.5 to 2 times greater than in nonsmoking locations. Median values for RSP were 12 μg m^{-3} higher in smoking homes than in nonsmoking homes (63 μg m^{-3} vs 51 μg m^{-3}). A corresponding increase in the ETS particle concentration of 10 μg m^{-3} (11 μg m^{-3} vs 1 μg m^{-3}) indicated that the vast majority of the increase in RSP levels in smoking homes was due to the additional contribution of ETS particles.

Assuming a breathing rate of 0.65 m^3 h^{-1}, an average level of respiration calculated for "awake" females (Holcomb 1993), housewives exposed to the median levels found in this study for nonsmoking households would be exposed to about 290 mg of RSP, 5.7 mg of ETS particles, and about 0.6 mg of nicotine in a year (Table 10). The corresponding exposures for individuals residing in smoking households found in this study were approximately 359 mg of RSP, 63 mg of ETS particles, and about 4.2 mg of nicotine in a year. For subjects living in smoking homes exposed to upper decile concentrations of RSP and ETS, representative of the most highly exposed housewives on this study, approximate annual exposures were equivalent to 883 mg of RSP, 501 mg of ETS particles, and 16 mg of nicotine. In this context, exposure may be defined as the "potential inhaled quantity" calculated as the product of the encountered concentration, the length of time subjected to such concentration, and the breathing rate maintained throughout the defined period.

The above calculations assume that subjects are exposed to these median (and upper decile) levels throughout the year. For comparison, a typical Spanish cigarette delivers about 15 mg of particles and 1 mg nicotine to the smoker, calculated from the mean yields of the top six selling brand-types in Spain. Putting these facts into perspective, housewives living in nonsmoking households would be exposed to less than 1 cigarette equivalent (CE) per year compared with approximately 4 CE for those living in smoking homes. These exposures are comparable with those found in Stockholm of less than 1 CE/y for nonsmoking house-

Table 11. Summary statistics of directly measured analytes for all housewives by age – Barcelona.

Analyte		Age range	Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
		20 - 34	0.50	8.4	2.5	1.3	1.5	11
Pre-cotinine	(ng mL^{-1})	35 - 49	0.50	5.2	14	1.2	0.50	27
		50 - 64	0.50	4.0	1.5	0.98	0.50	36
		20 - 34	0.50	1.9	0.96	0.75	0.50	6
Post-cotinine	(ng mL^{-1})	35 - 49	0.50	4.3	8.3	1.1	0.50	31
		50 - 64	0.50	2.9	1.4	0.86	0.50	36
		20 - 34	53	193	97	82	69	10
RSP	(µg m^{-3})	35 - 49	30	137	71	61	58	33
		50 - 64	33	110	69	57	51	39
		20 - 34	5.6	54	22	14	11	10
UVPM	(µg m^{-3})	35 - 49	2.6	40	15	8.6	6.4	32
		50 - 64	1.8	38	15	7.6	8.2	39
		20 - 34	5.2	20	14	11	10	9
FPM	(µg m^{-3})	35 - 49	2.3	20	12	6.7	5.2	30
		50 - 64	1.6	33	13	6.1	6.1	35
		20 - 34	2.7	70	27	8.8	6.6	10
SolPM	(µg m^{-3})	35 - 49	0.39	45	16	3.5	3.9	32
		50 - 64	0.14	47	15	2.9	3.6	39
		20 - 34	0.12	2.4	1.1	0.44	0.26	11
Nicotine	(µg m^{-3})	35 - 49	0.05	2.8	0.90	0.30	0.19	33
		50 - 64	0.04	2.0	0.78	0.33	0.24	38
		20 - 34	0.04	1.8	0.53	0.23	0.21	11
3-Ethenylpyridine	(µg m^{-3})	35 - 49	0.05	1.7	0.56	0.23	0.20	33
		50 - 64	0.04	1.6	0.54	0.27	0.25	38

holds and between 6 and 9 CE/y for smoking households (Phillips et al. 1996). However, care should be exercised when making comparisons based upon CEs, since the quantities of particles and nicotine delivered from an "average" cigarette may vary dramatically from country to country. CEs calculated for the Stockholm study were based on a delivery of 10 mg particles and 0.9 mg nicotine to the smoker, which were lower than the yields used for CE calculations in this study.

Annual exposures based upon upper decile concentrations of ETS particles and nicotine for housewives living in smoking households equate to between 16 and 33 CE/y.

CEs have been calculated using both SolPM and nicotine measurements throughout this publication. Where single figures have been quoted, the CE values calculated using both SolPM and nicotine measurements were the same.

Exposure difference by age: Summary analytical results by age are presented in Table 11. On examination of the exposure concentrations to RSP, ETS particles, and nicotine, there was a slight indication that the youngest age group, 20 to 34 y of age, may have been the most exposed. Saliva cotinine measurements taken after the monitoring period were not indicative of this trend, measured levels for each age group falling below the LOQ. This may indicate the poor reliability of using cotinine measurements, at the current LOQ, as a means for assessing ETS exposure.

Working subjects: Exposures at work and outside the workplace

In Barcelona, during the course of recruitment, it became apparent that the number of nonsmoking workplaces available for study was extremely low. This was reflected by the poor recruitment of subjects into Cells 4 and 6 with only 10% and 17% of the respective targeted numbers obtained, despite a significant amount of additional time being devoted to recruitment for these cells. As a consequence, comparison of data to provide meaningful analysis between individual cells

Table 12. Analytical statistics for working subjects based on their household smoking status. Home measurements – Barcelona.

Analyte		Cell *	Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
RSP	($\mu g\ m^{-3}$)	3 & 4	36	160	95	79	85	28
		5 & 6	28	105	59	49	40	40
UVPM	($\mu g\ m^{-3}$)	3 & 4	5.3	68	28	18	23	28
		5 & 6	2.0	29	11	5.8	4.3	40
FPM	($\mu g\ m^{-3}$)	3 & 4	5.0	69	28	18	25	28
		5 & 6	2.0	33	11	5.7	4.2	40
SolPM	($\mu g\ m^{-3}$)	3 & 4	1.5	99	35	14	21	28
		5 & 6	0.68	24	11	2.8	2.2	40
Nicotine	($\mu g\ m^{-3}$)	3 & 4	0.14	4.4	1.8	0.83	0.86	28
		5 & 6	0.06	0.61	0.48	0.21	0.17	41
3-Ethenylpyridine	($\mu g\ m^{-3}$)	3 & 4	0.15	2.0	0.96	0.58	0.63	28
		5 & 6	0.06	0.48	0.26	0.14	0.09	41

* Cells 3 & 4 - smoking households; Cells 5 & 6 - nonsmoking households.

Table 13. Analytical statistics for working subjects based on their workplace smoking status. Work measurements – Barcelona.

Analyte		Cell *	Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
RSP	($\mu g\ m^{-3}$)	3 & 5	47	197	112	94	94	60
		4 & 6	37	90	58	55	52	8
UVPM	($\mu g\ m^{-3}$)	3 & 5	5.7	87	46	26	29	60
		4 & 6	2.7	25	13	8.5	8.9	8
FPM	($\mu g\ m^{-3}$)	3 & 5	5.7	99	48	27	30	60
		4 & 6	2.5	26	13	8.8	10	8
SolPM	($\mu g\ m^{-3}$)	3 & 5	1.3	131	64	23	37	60
		4 & 6	0.39	28	11	2.7	2.6	8
Nicotine	($\mu g\ m^{-3}$)	3 & 5	0.43	9.0	4.1	2.3	2.4	61
		4 & 6	0.12	2.0	0.91	0.56	0.71	8
3-Ethenylpyridine	($\mu g\ m^{-3}$)	3 & 5	0.16	2.8	1.5	1.0	1.1	61
		4 & 6	0.12	1.7	0.63	0.37	0.29	8

* Cells 3 & 5 - smoking workplace; Cells 4 & 6 - nonsmoking workplace.

was not advisable due to the inadequate numbers of subjects recruited. Hence, cell data have been combined to provide summary analytical results according to smoking or nonsmoking environments. These are presented in Table 12 for the home (outside the workplace) and Table 13 for the workplace. In this instance, comparison of cotinine levels between environments was not possible due to the combination of cells to provide "environment" information.

Median levels of RSP, nicotine, and ETS particles were found to be higher in smoking environments than in nonsmoking environments. RSP levels in both the smoking home and the smoking workplace were approximately double those found in the corresponding nonsmoking environments. The highest median concentration of ETS particles (37 $\mu g\ m^{-3}$) was found within the smoking workplace and was about 14 times that of the nonsmoking workplace (2.6 $\mu g\ m^{-3}$). A breathing rate of 0.85 $m^3\ h^{-1}$, an average of the breathing rates for "awake" males (1.05 $m^3\ h^{-1}$) and females (0.65 $m^3\ h^{-1}$) reported by Holcomb (1993), was used for calculating exposures for working subjects where the concentration data generated was not sex dependent. This value of 37 $\mu g\ m^{-3}$, assuming this average breathing rate, equates to a work exposure of 0.25 mg/d based upon a mean time of 7.8 h spent in this environment.

Table 14. Analytical statistics for working subjects based on their home smoking status. Time weighted average values – Barcelona.

Analyte		Cell *	Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
Pre-cotinine	(ng mL^{-1})	3 & 4	0.50	8.3	3.2	1.8	1.6	22
		5 & 6	0.50	3.6	1.8	1.2	1.3	36
Post-cotinine	(ng mL^{-1})	3 & 4	0.50	4.5	2.5	1.7	1.8	24
		5 & 6	0.50	3.4	1.8	1.3	1.4	31
RSP	(μg m^{-3})	3 & 4	54	146	98	90	100	28
		5 & 6	36	115	74	64	60	40
UVPM	(μg m^{-3})	3 & 4	9.0	55	31	26	27	28
		5 & 6	4.3	43	23	14	14	40
FPM	(μg m^{-3})	3 & 4	9.2	55	32	26	29	28
		5 & 6	4.3	48	23	15	15	40
SolPM	(μg m^{-3})	3 & 4	9.0	83	41	30	34	28
		5 & 6	1.7	79	28	12	16	40
Nicotine	(μg m^{-3})	3 & 4	0.51	3.9	2.1	1.5	1.7	28
		5 & 6	0.19	3.7	1.8	0.98	1.1	41
3-Ethenylpyridine	(μg m^{-3})	3 & 4	0.36	2.0	1.1	0.86	0.98	28
		5 & 6	0.13	1.3	0.65	0.45	0.45	41

* Cells 3 & 4 - smoking households; Cells 5 & 6 - nonsmoking households.

Corresponding median ETS particle concentrations outside the workplace were 2.2 μg m^{-3} for subjects living in nonsmoking homes and 21 μg m^{-3} (9.5 times higher) for those living in smoking homes. Hence, workers who live in smoking households are exposed to 0.28 mg ETS particles per d based upon a mean time of 15.6 h spent outside the workplace. The level of work exposure, accrued over a much shorter period of time, was higher than that found for housewives living in smoking homes (11 μg m^{-3}) which was equivalent to an exposure of 0.17 mg/d. These values are significantly higher than those reported for Stockholm (Phillips et al. 1996), median ETS particle concentrations being at least 15 times greater for those subjects living or working in smoking environments and at least 3 times higher for those subjects living or working in nonsmoking environments. These levels are close to those reported by Holcomb (1993) of between 0.06 and 0.1 mg per d calculated from literature values of ETS concentrations. Repace and Lowrey (1985) estimated levels of exposure between 1.43 mg and 14.3 mg/d of RSP from ETS during the 1980s.

Median nicotine levels determined for working subjects in the workplace were higher for those in a smoking environment (2.4 μg m^{-3}) than a nonsmoking environment (0.71 μg m^{-3}). This median value of 2.4 μg m^{-3} for smoking workplaces was lower than those found by Hammond et al. (1995) who reported median levels of 9.1 μg m^{-3} for closed offices (small rooms with low occupancy) and 8.6 μg m^{-3} for open offices (multiple occupancy of open plan area with or without partitions) where smoking was permitted. However, this level of exposure exceeds that quoted by Repace and Lowrey (1993) who estimated that a workplace nicotine exposure of 2.3 μg m^{-3}, over a working period of 40 y, presented a lung cancer risk of 3 in 10 000, which in the U.S. is high enough to provoke the intervention of regulatory agencies to reduce levels. Median levels quoted by Hammond et al. (1995) for open offices where smoking is restricted (1.3 μg m^{-3}) or banned (0.3 μg m^{-3}) are comparable with the median level found for nonsmoking workplaces in this study.

Outside the workplace, corresponding nicotine exposure concentrations of 0.86 μg m^{-3} and 0.17 μg m^{-3} were found for smoking and nonsmoking households, respectively. Although median nicotine levels were more than 3 times lower in nonsmoking workplaces than smoking workplaces, the levels found were comparable with those apparent outside the workplace for subjects living in smoking households (0.71 μg m^{-3} vs 0.86 μg m^{-3}). This median concentration apparent in the "nonsmoking" workplace highlights the possibility of indistinct segregation of smoking and nonsmoking workplaces in Barcelona.

Table 14 shows summary analytical time weighted results for subjects according to smoking or nonsmoking home environments, irrespective of the smoking status of their workplace. Each subject's mean

Table 15. Calculated annual exposures to RSP, ETS particles, and nicotine for working subjects – Barcelona.

Environment	Annual exposure (mg) RSP	ETS particles[a]	Nicotine	Cigarette equivalents[b]
		Median levels		
Smoking home	512	126	5.2	5.2 - 8.4
Nonsmoking home	241	13	1.0	0.9 - 1.0
Smoking work	134	53	3.4	3.4 - 3.5
Nonsmoking work	74	3.7	1.0	0.2 - 1.0
		Upper decile levels		
Smoking home	963	596	26	26 - 40
Nonsmoking home	632	144	3.7	3.7 - 9.6
Smoking work	281	187	13	12 - 13
Nonsmoking work	129	40	2.9	2.7 - 2.9

[a] Estimated using solanesol measurements (SolPM).
[b] Calculated using both ETS particle and nicotine measurements.

Table 16. Saliva cotinine levels for working subjects by sex – Barcelona.

Analyte		Sex	Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
Pre-cotinine	(ng mL^{-1})	M	0.50	3.0	1.8	1.2	1.2	30
		F	0.50	6.0	2.9	1.7	1.7	28
Post-cotinine	(ng mL^{-1})	M	0.50	3.8	2.1	1.4	1.3	28
		F	0.50	4.0	2.1	1.5	1.7	27

exposure concentration was calculated by combining the "home" and "work" measurements in proportion with the time spent in each environment. Median pre- and post-monitoring saliva cotinine levels were slightly elevated for those subjects living in smoking households (1.6 and 1.8 ng mL^{-1} vs 1.3 and 1.4 ng mL^{-1}), although not as marked as other indicators for ETS exposure. Working subjects living in smoking homes had the highest median time weighted nicotine exposure (1.7 μg m^{-3}) which was comparable with the average exposure level of 1.61 μg m^{-3} reported by Ogden et al. (1993) for employed women in the U.S. living in smoking households.

Annual exposures calculated from determined median and upper decile concentrations of RSP, ETS particles, and nicotine in smoking and nonsmoking home and work environments are presented in Table 15. These values are calculated assuming a 35-h working week and a 48-week working year, with the remaining time spent outside the workplace. Therefore, based on median levels, subjects working in nonsmoking environments or living in nonsmoking households would be exposed to a maximum of one CE/y in each location. Subjects living in smoking households would be exposed to between 5.2 and 8.4 CE/y outside the workplace and subjects employed in smoking workplaces would be exposed to between 3.4 and 3.5 CE/y whilst at work. These exposures where smoking takes place are at least 10 times higher than those reported for Stockholm (Phillips et al. 1996).

Upper decile exposures based upon nicotine were approximately 3 and 4 times higher for nonsmoking and smoking environments, respectively. Corresponding upper decile exposures based upon ETS particles were approximately 11 and 4 times higher, respectively. Although the contribution to overall exposures from the workplace was considered significant, Tables 14 and 15 show that the major contribution to ETS exposure comes from living in a household where smoking takes place.

Comparison of exposures in working subjects by age and sex distribution: Summary analytical results by age and sex are presented in Tables 16 through 21. Median saliva cotinine levels determined for female subjects

Table 17. Analytical statistics for working subjects by sex. Home measurements – Barcelona.

Analyte		Sex	Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
RSP	($\mu g\ m^{-3}$)	M	28	103	58	49	45	35
		F	33	159	90	74	85	33
UVPM	($\mu g\ m^{-3}$)	M	2.0	33	11	6.4	4.3	35
		F	3.9	68	24	14	11	33
FPM	($\mu g\ m^{-3}$)	M	1.9	34	12	6.3	4.5	35
		F	3.9	67	24	14	9.3	33
SolPM	($\mu g\ m^{-3}$)	M	0.63	38	11	3.2	2.4	35
		F	1.3	101	31	10	7.1	33
Nicotine	($\mu g\ m^{-3}$)	M	0.07	2.7	0.98	0.28	0.22	36
		F	0.08	2.8	1.1	0.48	0.50	33
3-Ethenylpyridine	($\mu g\ m^{-3}$)	M	0.06	1.3	0.38	0.18	0.15	36
		F	0.06	1.9	0.73	0.36	0.28	33

Table 18. Analytical statistics for working subjects by sex. Work measurements – Barcelona.

Analyte		Sex	Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
RSP	($\mu g\ m^{-3}$)	M	36	171	93	78	76	35
		F	48	211	119	100	93	33
UVPM	($\mu g\ m^{-3}$)	M	4.9	75	32	19	23	35
		F	5.8	92	53	28	40	33
FPM	($\mu g\ m^{-3}$)	M	5.2	86	35	20	24	35
		F	5.4	99	53	29	42	33
SolPM	($\mu g\ m^{-3}$)	M	1.1	100	38	13	18	35
		F	1.2	157	78	25	59	33
Nicotine	($\mu g\ m^{-3}$)	M	0.29	7.0	3.3	1.7	2.1	36
		F	0.43	9.1	4.2	2.3	2.3	33
3-Ethenylpyridine	($\mu g\ m^{-3}$)	M	0.11	2.6	1.2	0.71	0.81	36
		F	0.21	2.8	1.6	1.1	1.5	33

Table 19. Saliva cotinine levels for working subjects by age – Barcelona.

Analyte		Age range	Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
Pre-cotinine	($ng\ mL^{-1}$)	20 - 34	0.50	6.3	2.6	1.9	1.7	18
		35 - 49	0.50	4.3	1.8	1.2	1.1	29
		50 - 64	0.50	8.5	3.3	1.5	1.1	11
Post-cotinine	($ng\ mL^{-1}$)	20 - 34	0.50	3.3	2.2	1.6	1.7	15
		35 - 49	0.50	3.9	2.0	1.4	1.4	27
		50 - 64	0.50	3.9	2.2	1.4	1.6	12

(Table 16) were higher than the corresponding values found for male subjects (pre- 1.7 ng mL^{-1} vs 1.2 ng mL^{-1}; post- 1.7 ng mL^{-1} vs 1.3 ng mL^{-1}). Median exposures to RSP and ETS particles for workers outside the workplace (Table 17) were also higher for women than for men with RSP approximately twofold higher (85 $\mu g\ m^{-3}$ vs 45 $\mu g\ m^{-3}$) and ETS particles approximately 3 times higher (7.1 $\mu g\ m^{-3}$ vs 2.4 $\mu g\ m^{-3}$). Median nicotine levels were also elevated. In the workplace (Table 18), the same trend was evident, with all median concentrations being higher than those determined outside the workplace. For women, RSP was elevated by

Table 20. Analytical statistics for working subjects by age. Home measurements – Barcelona.

Analyte		Age range	Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
		20 - 34	29	137	79	61	73	21
RSP	(μg m^{-3})	35 - 49	34	157	74	60	51	32
		50 - 64	29	127	62	53	50	14
		20 - 34	2.2	33	17	9.9	11	21
UVPM	(μg m^{-3})	35 - 49	2.4	54	16	8.3	6.4	32
		50 - 64	2.8	38	19	10	7.1	14
		20 - 34	2.3	38	18	9.6	9.1	21
FPM	(μg m^{-3})	35 - 49	2.1	60	17	8.0	6.2	32
		50 - 64	2.9	38	19	11	7.9	14
		20 - 34	0.26	55	19	5.2	5.6	21
SolPM	(μg m^{-3})	35 - 49	0.79	94	21	5.1	3.6	32
		50 - 64	0.73	49	21	6.3	4.6	14
		20 - 34	0.08	3.2	1.4	0.43	0.29	21
Nicotine	(μg m^{-3})	35 - 49	0.06	2.5	0.83	0.31	0.23	33
		50 - 64	0.08	2.3	0.89	0.37	0.32	14
		20 - 34	0.06	1.4	0.51	0.26	0.23	21
3-Ethenylpyridine	(μg m^{-3})	35 - 49	0.06	1.5	0.50	0.22	0.18	33
		50 - 64	0.06	1.8	0.64	0.29	0.20	14

Table 21. Analytical statistics for working subjects by age. Work measurements – Barcelona.

Analyte		Age range	Lower decile	Upper decile	Arithmetic mean	Geometric mean	Median	Number of subjects
		20 - 34	43	236	130	109	119	21
RSP	(μg m^{-3})	35 - 49	40	170	97	78	87	32
		50 - 64	65	145	92	86	86	14
		20 - 34	5.7	138	59	34	47	21
UVPM	(μg m^{-3})	35 - 49	4.5	70	35	16	18	32
		50 - 64	8.1	69	34	25	27	14
		20 - 34	6.5	160	65	37	48	21
SolPM	(μg m^{-3})	35 - 49	4.4	73	32	17	18	32
		50 - 64	10	81	38	28	28	14
		20 - 34	2.6	231	92	33	63	21
FPM	(μg m^{-3})	35 - 49	1.1	112	42	11	14	32
		50 - 64	1.0	71	40	18	37	14
		20 - 34	0.60	8.2	4.1	2.8	4.3	21
Nicotine	(μg m^{-3})	35 - 49	0.33	9.3	4.1	1.7	1.5	33
		50 - 64	0.28	4.4	2.6	1.6	2.1	14
		20 - 34	0.46	2.8	1.9	1.5	1.8	21
3-Ethenylpyridine	(μg m^{-3})	35 - 49	0.12	2.7	1.1	0.64	0.70	33
		50 - 64	0.13	2.0	1.1	0.76	1.1	14

approximately 20% (93 μg m^{-3} vs 76 μg m^{-3}), ETS particles were 3 times higher than for men (59 μg m^{-3} vs 18 μg m^{-3}), and nicotine levels were comparable (2.3 μg m^{-3} vs 2.1 μg m^{-3}). This median ETS particle concentration of 59 μg m^{-3} determined for female workers equates to an annual exposure, assuming a breathing rate of 0.65 m^3 h^{-1}, of 4.3 CE whilst at work.

For working subjects outside the workplace (Table 20), similar median exposure concentrations for RSP (50 - 73 μg m^{-3}), ETS particles (3.6 - 5.6 μg m^{-3}), and

nicotine (0.23 - 0.32 μg m^{-3}) were apparent across the age ranges investigated. There was, however, a slight indication that the younger age group (20 - 34 y) may have been more highly exposed. Median cotinine levels determined for each of the age ranges investigated (Table 19) were also indicative of this trend. When comparing the age ranges for employed subjects at work (Table 21), the above trend is more pronounced with median levels of RSP, ETS particles, and nicotine for the 20 - 34 y age group being 1.4, 1.7, and 2 times higher than the next highest median value reported.

Income levels and exposure in working subjects: As part of the last visit survey, subjects were required to indicate the level of annual household income within stated earning brackets from 2 million pesetas (pts) or below, increasing to 20 million pts and above. There were insufficient numbers in any of the earning brackets above 6 million pts to enable comparison of ETS exposure with household income at this level. Summary statistics for the levels of ETS exposure in the household income brackets of up to 2 million pts, between 2 million and 4 million pts, and between 4 million and 6 million pts were calculated. Although definite conclusions could not be drawn from the data, there was a strong indication that workers from households with incomes in the lowest bracket may have been exposed to higher levels of ETS, especially in their place of work.

Subjective comparisons of ETS exposure

Subjective assessments of ETS exposure were made by subjects immediately after sampling for 24 h and again on their return to the study centre (Figs. 6, 7, and 8). Housewives and workers outside the workplace made comparable assessments of their ETS exposures during the monitoring period and, in the workplace, employed subjects considered themselves to be the most highly exposed. These subjective assessments were consistent with measured levels, the highest exposure concentrations having been measured for workers within the workplace. However, working subjects are actually more exposed outside the workplace since the majority of the time was not spent at work, indicating that subjects do not take account of the time spent in any one location when making a subjective assessment of exposure. Questionnaire data does, however, have some significance when used in conjunction with measured concentrations. Riboli et al. (1990) also concluded that, when appropriately questioned, nonsmoking subjects (women in this instance) can provide estimates of ETS exposure which are good indicators of their biochemically measured exposure levels.

It is interesting to note that information from subjects' diaries, completed during the monitoring periods and last visit survey questionnaires, indicates that approximately 17% of all subjects living or working in smoking environments did not see or smell any tobacco smoke during the monitoring period. Also apparent from this information was the fact that about 27% of all subjects living or working in nonsmoking environments did note smoking during the monitoring period. This might indicate an ad hoc policy on smoking, particularly in the workplace.

In the single monitor study, housewives living in nonsmoking homes who reported the presence of smoking during their monitoring periods were exposed to approximately 10 times the median levels of ETS particles (6.5 μg m^{-3} vs 0.67 μg m^{-3}) and approximately 3 times the median levels of nicotine (0.29 μg m^{-3} vs 0.10 μg m^{-3}) determined for those who did not report smoking. As would be expected, RSP levels were also elevated (64 μg m^{-3} vs 45 μg m^{-3}). Conversely, housewives living in smoking households who did not report the presence of tobacco smoke during their monitoring periods were exposed to lower median concentrations of ETS particles (1.8 μg m^{-3} vs 17 μg m^{-3}), nicotine (0.23 μg m^{-3} vs 1.1 μg m^{-3}), and RSP (44 μg m^{-3} vs 75 μg m^{-3}) than those subjects who did report smoking. These elevations in the levels of ETS constituents where smoking was reported are consistent with those found for Stockholm (Phillips et al. 1996).

For the dual monitor study, subjects living in smoking homes who reported seeing or smelling smoking outside the workplace were again exposed to more ETS than those who did not. Median levels of ETS particles and nicotine were about 14 and 8 times higher, respectively, in this instance, with a corresponding twofold increase in RSP levels. Working subjects living in nonsmoking households were exposed to 5 and 3 times higher concentrations of ETS particles and nicotine at home and all other locations outside the workplace where smoking was reported to have occurred during the sampling period.

The number of subjects working in nonsmoking locations and the number of subjects working in smoking locations who did not report smoking were low. Hence, a valid comparison of exposure data was difficult to make. However, with the limited data available, indications were that the same pattern was evident. Personal exposures to ETS particles, nicotine, and RSP

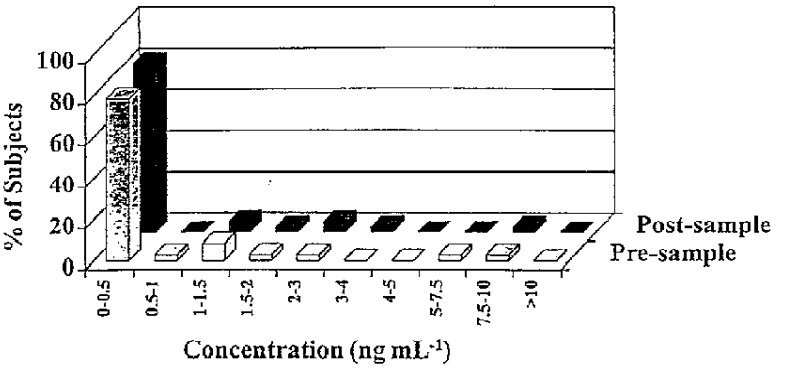

Fig. 9. Distribution of saliva cotinine concentrations. Nonsmoking homes (Cell 2) – Barcelona.

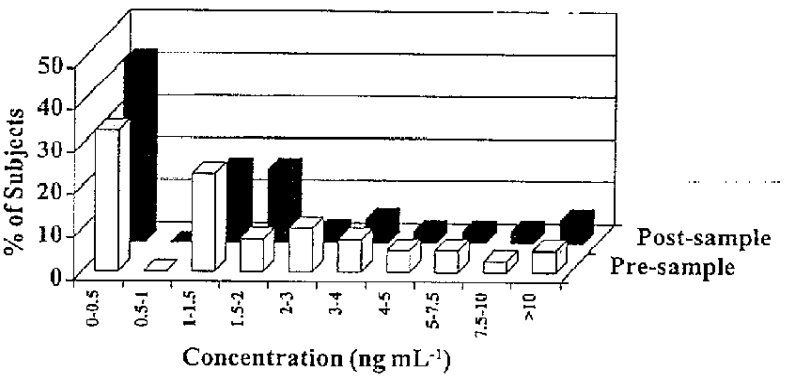

Fig. 10. Distribution of saliva cotinine concentrations. Smoking homes (Cell 1) – Barcelona.

appeared much higher, in both nonsmoking and smoking environments, where subjects had reported the presence of tobacco smoke when compared with subjects who had not. These findings lend weight to the accuracy and integrity of study questionnaires and diaries completed by subjects throughout the monitoring period during this study.

A nonsmoking workplace was defined by the absence of smoking co-workers within 30 m of a subject's workplace and was independent of any employer's smoking/nonsmoking policy. No account of the magnitude of smoking observed on any occasion has been made in the calculation of these values.

In addition to the documentation of observed smoking activity during the monitoring period, as part of the last visit survey, subjects were required to provide responses to a number of subjective questions regarding their exposure to ETS both prior to and during study participation. When asked which location was considered to be the highest contributor to overall ETS exposure, 50% of participant responses indicated this to be in a bar area. The next highest contributor, comprising 17% of participant responses, was considered to be the workplace. This level of response for bar areas, usually associated with high levels of ETS, again indicating that subjects associate high ETS concentrations with high levels of exposure irrespective of the length of time spent in any one environment.

Comparison of other measures of ETS exposure

Cotinine: Figures 9 and 10 show the distributions of pre- and post-monitoring saliva cotinine concentrations for housewives living in nonsmoking and smoking households, respectively. A greater proportion of higher saliva cotinine concentrations are clearly evident for those subjects residing in smoking households, which is consistent with the findings of NHANES III (Pirkle et al. 1996), where measured serum cotinine levels showed the same pattern. This finding would seem to suggest that this measurement may be of some use for assessing ETS exposure. However, when correlations of post-cotinine concentrations with more direct measures of ETS exposure were calculated, only weak relationships were apparent. The best correlation was found between the single monitor (housewife) study post-cotinine and FPM measurements with an R^2 value of 0.436. Correlations with SolPM, nicotine, and 3-EP had R^2 values of 0.194, 0.134, and 0.156, respectively. These findings further reinforce a belief that saliva cotinine measurements, using the current LOQ, are not a reliable marker for ETS exposure.

SolPM: SolPM, currently used as a tobacco specific marker, should provide the most accurate means of assessing ETS particles in air. However, with the current LOQ obtainable for the method, at low concentrations of ETS particles a greater proportion of SolPM analyses was below the LOQ than for either UVPM or FPM determinations. Occasionally, the nicotine levels corresponding with SolPM values below the LOQ were measurable. This may be due to contributions from background nicotine levels from a variety of sources, in particular desorption from surfaces and fabrics. Nicotine may also emanate from ventilation systems or by convection in air currents moving from one location to another.

At high concentrations, SolPM measurements may well be overestimating the levels of ETS particles present since elevated levels estimated using SolPM were in excess of those using either UVPM or FPM methods. On five occasions, ETS particle concentrations calculated from solanesol measurements exceeded the determined RSP levels. These higher levels or "overestimates" may be due to the use of factors established in a model room based on "fresh" cigarette smoke, and not a mixture of "aged" ETS particles from various tobacco products.

Direct comparison of SolPM exposure concentrations with UVPM measurements gave a good correlation ($R^2 = 0.933$) with a similar value apparent when correlated with FPM measurements ($R^2 = 0.920$).

Nicotine and 3-EP: Although the LOQs for the determination of nicotine and 3-EP are the same, the amounts of nicotine quantified during the study were higher than those for 3-EP. Approximately 13% of all nicotine measurements made during the study were below the LOQ compared with approximately 24% for 3-EP. The correlation between nicotine and 3-EP exposure concentrations was moderate with an R^2 value of 0.608. Correlations with measures for ETS particulate concentrations were also moderate, all R^2 values for comparisons with 3-EP being higher than for nicotine (0.662 and 0.507 vs SolPM, 0.751 and 0.542 vs FPM, 0.676 and 0.620 vs UVPM, respectively).

3-EP may be regarded as unique to tobacco smoke (Eatough et al. 1989), present in the ETS vapour phase at measurable concentrations, and is claimed to decay at rates similar to those of common gases such as carbon dioxide and carbon monoxide. These facts together with these findings may suggest that 3-EP is a better comparator for ETS exposure than nicotine, a conclusion similar to that drawn by Sterling et al. (1996), although an improved LOQ would be desirable to justify its use in preference to nicotine. In a recent study, Hodgson et al. (1996) examined the usefulness of 3-EP as a tracer to determine ETS contributions to overall volatile organic compounds in smoking environments, where again the need for an improved LOQ was highlighted. Nelson et al. (1992) also concluded that 3-EP was superior to nicotine as a vapour phase marker for ETS, and, if its decay characteristics were similar to that of the particulate phase as their findings in a test chamber suggest, it may be of use in predicting both gas and particulate phase compounds in ETS.

CONCLUSIONS

Recruited subjects

Using a segmentation system enabled the study to attract a subject sample closely resembling the population of Barcelona. Recruitment of subjects working in nonsmoking workplaces was found to be extremely difficult. This was probably attributable to the fact that, according to Catalunya state law, smoking is only prohibited in the following workplaces: health centres, child care centres (under 16 y), education centres, administration centres open to the public, theatres, cinemas, and establishments where food is prepared,

manipulated, transformed, or sold with the exception of those mainly devoted to food consumption.

Housewives

Median levels of RSP, ETS particles, nicotine, and 3-EP determined for housewives living in nonsmoking homes were the lowest found in this study. The levels were between 3 and 8 times higher than those found for similar housewives in Stockholm (Phillips et al. 1996). For housewives living in smoking homes, median levels for ETS particles and nicotine were both less than those determined for equivalent housewives in Stockholm. However, the corresponding median RSP level was 62% higher in Barcelona than in Stockholm (63 $\mu g\ m^{-3}$ vs 39 $\mu g\ m^{-3}$). Annualized exposures based on upper decile levels for those residing in smoking homes, the most highly exposed housewives in Barcelona, showed that these subjects would potentially inhale or "receive" 883 mg RSP, 501 mg of which being attributable to ETS particles, and 16 mg nicotine. This equates to between 16 and 33 CE/y.

Working subjects

A combination of smoking cells, and similarly nonsmoking cells, was undertaken to enable the comparison of exposures where recruitment was extremely low. The highest concentrations, based on medians, were found for subjects in smoking workplaces. Levels of 94 $\mu g\ m^{-3}$ RSP, 37 $\mu g\ m^{-3}$ ETS particles, and 2.4 $\mu g\ m^{-3}$ nicotine were recorded, and were 6, 33, and 12 times higher than for equivalent workers in Stockholm (Phillips et al. 1996). This median nicotine concentration within the workplace, over a working period of 40 y, has been estimated to present a lung cancer risk of 3 in 10 000 (Repace and Lowrey 1993).

If the length of time spent in each location (inside/outside the workplace) is taken into consideration, the highest exposures were for workers outside the workplace living in smoking households. Annually, these subjects would be exposed to (or potentially inhale) 512 mg RSP, 126 mg ETS particles, and 5.2 mg nicotine outside the workplace, which equates to between 5.2 and 8.4 CE. Total annualized exposures may be estimated by the combination of exposures calculated for workers both inside and outside the workplace. Based on median levels, workers employed in smoking workplaces and living in smoking households would be exposed to between 8.6 and 12 CE/y, at least 6 times higher than for workers living and working in nonsmoking environments (1.1 - 2 CE). The most highly exposed subjects in this study, based upon upper decile concentrations for those living in smoking households and working in smoking workplaces, would be exposed to between 38 and 53 CE in a year.

Saliva cotinine

A concentration of 25 ng mL^{-1} was again chosen as the cut-off point for distinguishing between smokers and nonsmokers. In a recent publication, Sterling et al. (1996) indicated a range quoted from reviewed literature of between 20 and 100 ng mL^{-1}, opting for the higher level cut-off at 100 ng mL^{-1}. Sterling et al. believe a possible explanation for high cotinine levels found for two of their subjects (32.9 and 79.6 ng mL^{-1}) was dietary intake of nicotine because the corresponding measurements of particulate and vapour phase components were comparable with the levels found for subjects with saliva cotinine levels below 10 ng mL^{-1}. These findings suggest that saliva cotinine levels may not be a good marker for ETS exposure with such a wide variation in measured results apparent. The NHANES III report (Pirkle et al. 1996) indicated, using various regression analyses, that the geometric mean contribution of dietary intake to serum cotinine levels was less than 0.02 ng mL^{-1}. It is reasonable to assume similar values for saliva cotinine levels. In a recent examination of the use of cotinine as a biomarker to infer ETS exposure for nonsmokers (Chappell and Gratt 1996), the possibility of diet as a significant source of nicotine was highlighted since the estimated dose of nicotine from ETS exposure was less than the total dose estimated from cotinine concentrations. Hence, an overestimation of ETS exposure may result from the use of saliva cotinine levels to predict air nicotine concentrations. The dietary contribution of nicotine from tea was highlighted by these authors, which may be significant in countries where tea drinking is popular and widespread. For the purpose of this study, the dietary contribution of nicotine has been ignored, measurements having been used as a guide to smoking status and for providing additional exposure information.

For working subjects, overall median saliva cotinine levels were 1.5 ng mL^{-1} for both pre- and post-sampling measurements. For housewives living in smoking households, median saliva cotinine levels were 1.3 and 1.1 ng mL^{-1} for pre- and post-measurements, respectively, with corresponding levels for those living in nonsmoking households below the LOQ. Median saliva cotinine levels for working subjects living in smoking households were 1.6 and 1.8 ng mL^{-1} for pre- and post-

measurements, respectively, compared with 1.3 and 1.4 ng mL^{-1}, respectively, for those living in non-smoking households. Overall, the saliva cotinine levels found in this study for subjects living and working in Barcelona are up to 2 times the equivalent levels found in the population of Stockholm (Phillips et al. 1996).

Questionnaires

Subjective assessments of ETS exposure made by subjects immediately after sampling for 24 h and again on their return to the study centre were consistent with measured levels. The measured exposure concentrations for workers outside the workplace and for housewives were comparable, which was mirrored by similar distributions for subjective assessments of exposure. A greater proportion of subjects indicated having been more highly exposed in the workplace where the highest concentrations, but not necessarily the highest exposures, were measured. This suggests that subjects did not take account of the time spent in any one location when making a subjective assessment of overall exposure. Questionnaire data would appear to have more significance when used in conjunction with measured concentrations, a finding similar to that of Riboli et al. (1990).

Estimation of ETS

UVPM, FPM, and SolPM methods were used to estimate ETS particle concentrations, each giving a different value. There was an indication that inconsistent relative values were produced depending upon the magnitude of ETS particles collected. Further work on the use of both solanesol and 3-EP as overall markers for ETS concentrations should be investigated.

Exposure calculations

During the composition of this paper and others in the series, comparing daily or annualized exposures with other values cited in published literature was considered. There appears to be only limited agreement on the use of sampling times (Ogden et al. 1996), average inhalation volumes (Arundel et al. 1988), and cigarette equivalents (Holcomb 1993) without even considering the use of means, medians, or most highly exposed as part of any calculation. This and other studies should provide sufficient data to enable subsequent calculations to be made.

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